Dissolution and Analysis of Nuclear Fuels and Targets

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Introduction

The Advanced Fuel Cycle Initiative (AFCI) is developing advanced proliferation-resistant technologies that allow the safe and economical disposal of waste from nuclear reactors. A critical element of this initiative is the separation and disposition of key radionuclides. **The first step in the separation scheme is to dissolve the irradiated fuel.**

The U.S. Reduced Enrichment for Research and Test Reactors (RERTR) program is working to limit the use of high-enriched uranium (>20% ^{235}U) by substituting low-enriched uranium (<20% ^{235}U) fuel and targets. Technetium-99m, the daughter of ^{99}Mo, is the most commonly used medical radioisotope in the world. Currently, most of the world's supply of ^{99}Mo is produced by fissioning HEU in targets. An LEU target must contain five times the uranium as an HEU target; uranium metal foil maximizes the U density in the target. **The first step in the ^{99}Mo recovery is to dissolve the irradiated target.**

Challenges

1. Dissolve irradiated oxide nuclear fuel in the **low acid** conditions required for the aqueous separation flow sheet.
2. Dissolve irradiated U metal targets (for ^{99}Mo production) in the **low base** conditions required for ^{99}Mo purification and recovery.

Our Approach

1. Dissolve the oxide fuel at relatively **high temperature** to increase the solubility and increase the dissolution rate.
2. Dissolve U metal targets using **$KMnO_4$ to oxidize** the U metal to uranyl hydroxide. This oxidation leads to the release of Mo into the solution.

Results and Conclusions

1. The irradiated oxide nuclear fuel was successfully dissolved under low acid conditions. However, a Pu-rich zirconium molybdate phase formed during storage of the dissolved fuel
2. The irradiated U metal targets were successfully digested under low base conditions. The use of $KMnO_4$ as an oxidant is shown to affect the ^{99}Mo yield.

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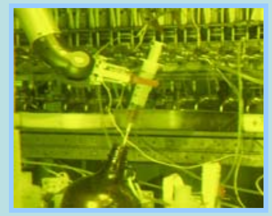
Irradiated Fuel Dissolution

Dissolver



Dissolved Fuel

Fuel in Cladding

Residue

Dissolved Fuel Composition

Element	g/L	Element	g/L	Element	g/L
U	**361**	Sb	**0.42**	Np	**0.17**
Pu	**2.68**	La	**0.39**	Mo	**0.16**
Gd	**2.64**	Am	**0.31**	Y	**0.15**
Nd	**1.39**	Sm	**0.27**	Rb	**0.11**
Ba	**1.36**	Sr	**0.26**	Eu	**0.036**
Ru	**0.78**	Tc	**0.26**		
Ce	**0.67**	Zr	**0.21**		
Cs	**0.63**	Rh	**0.17**		

Residue Composition

Element	wt%	Element	wt%
Mo	**20.2**	Cd	**0.18**
Pu	**13.6**	Tc	**0.16**
Zr	**12.8**	Sr	**0.053**
U	**1.41**	Cs	**0.046**
Te	**1.29**	Eu	**0.000072**
Ba	**0.44**	Am	**0.00028**
Ag	**0.23**		
Ru	**0.20**		

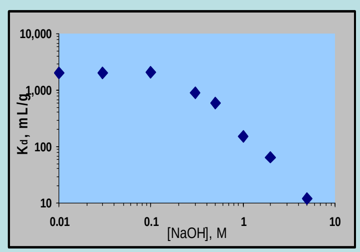
Uranium Metal Target Digestion

$$U + 2KMnO_4 + 2H_2O \Leftrightarrow UO_2(OH)_2 + 2MnO_2 + 2KOH$$

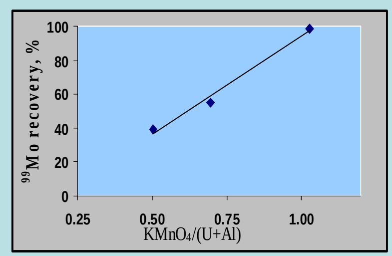
$$Al + 3\,H_2O \Leftrightarrow \underline{Al(OH)_3}\ \text{(unfilterable)} + 3/2\,H_2$$

Al and Mn coprecipitate to form a filterable solid

Digester



^{99}Mo purification/recovery efficiency is better under low base conditions



^{99}Mo recovery depends on the amount of $KMnO_4$ present.